NSLS-II Beamline Report for SAC Review of Beamlines

Beamline: **Coherent Hard *X-ray (CHX) Beamline*, *11-ID***
Report Date: **March 2019**

Introduction

The Coherent Hard X-ray (CHX) beamline at 11-ID was designed and constructed to provide hard x-ray coherent scattering capabilities with world-leading coherent flux and temporal resolution. The instrument offers capabilities for both small- and wide-angle X-ray scattering (SAXS, WAXS) including simultaneous SAXS-WAXS. Consequently, the beamline can access structural information on length scales covering Ångstroms to microns. The beamline optics provides a suite of technical solutions for achieving high spatial and temporal stability while offering the required tunability to match the optical resolution with the pixel sizes of the fastest state-of-the-art pixel array detectors available. The beamline is optimized for studies of dynamical phenomena using X-ray Photon Correlation Spectroscopy (XPCS) and other coherent scattering techniques such as X-ray Speckle Visibility Spectroscopy (XSVS). The time scales achieved on a routine basis in XPCS cover many orders of magnitude, from ~100 μs (9 kHz) to hours. In special cases, with strong scatterers, XSVS can be used to access even shorter time scales approaching 1 μs.

The CHX beamline was developed as one of the seven NSLS-II Project beamlines, funded through DOE Basic Energy Sciences (BES). The CHX Beamline received the first synchrotron light in Nov 2014, performed the first science commissioning user experiments during the 2015-2 cycle and received approval for general user operation in September 2016. The general user program has been aggressively ramped-up with a current experimental beam time usage of ~80%.

Beamline Overview

The research program of the CHX beamline takes advantage of the extraordinary opportunities provided by the NSLS-II high-brightness source to address new challenges in materials science that impact novel energy applications and advanced manufacturing techniques such as 3D printing or self-assembly. As systems of technological interest become smaller, more complex and nano-structured, non-stationary and non-equilibrium effects often associated with their temporal and spatial heterogeneities become increasingly important. This reality was recognized in a recent report by the Department of Energy Sciences Advisory Committee [1]. One of the ``Five Grand Challenges for Science and Imagination" identified in their report is summarized by the far-reaching question: ``How do we characterize and control matter away - especially very far away - from equilibrium?”. The capabilities of the CHX instrument aim at providing experimental tools and experimental methods that address this Grand Challenge.

The Coherent Hard X-ray (CHX) beamline at NSLS-II is dedicated to studies of nanometer-scale dynamics in materials using X-ray Photon Correlation Spectroscopy (XPCS), and to other experimental methods enabled by bright, coherent, X-ray beams. XPCS is based on measuring time correlation functions of the speckle fluctuations that occur when a coherent X-ray beam is scattered by a heterogeneous sample. For some recent reviews of XPCS, see for e.g. [2-4] and references therein. Figure 1a shows a speckle pattern recorded at the CHX beamline.

The key quantity that enables XPCS experiments is the source brightness. This determines the flux of coherent X-ray photons and ultimately the signal-to-noise ratio (SNR) of the measured correlation functions. The tremendous source brightness at the NSLS-II now enables XPCS experiments covering time scales orders of magnitudes faster than what was achievable only a few years ago (Figure 1b)

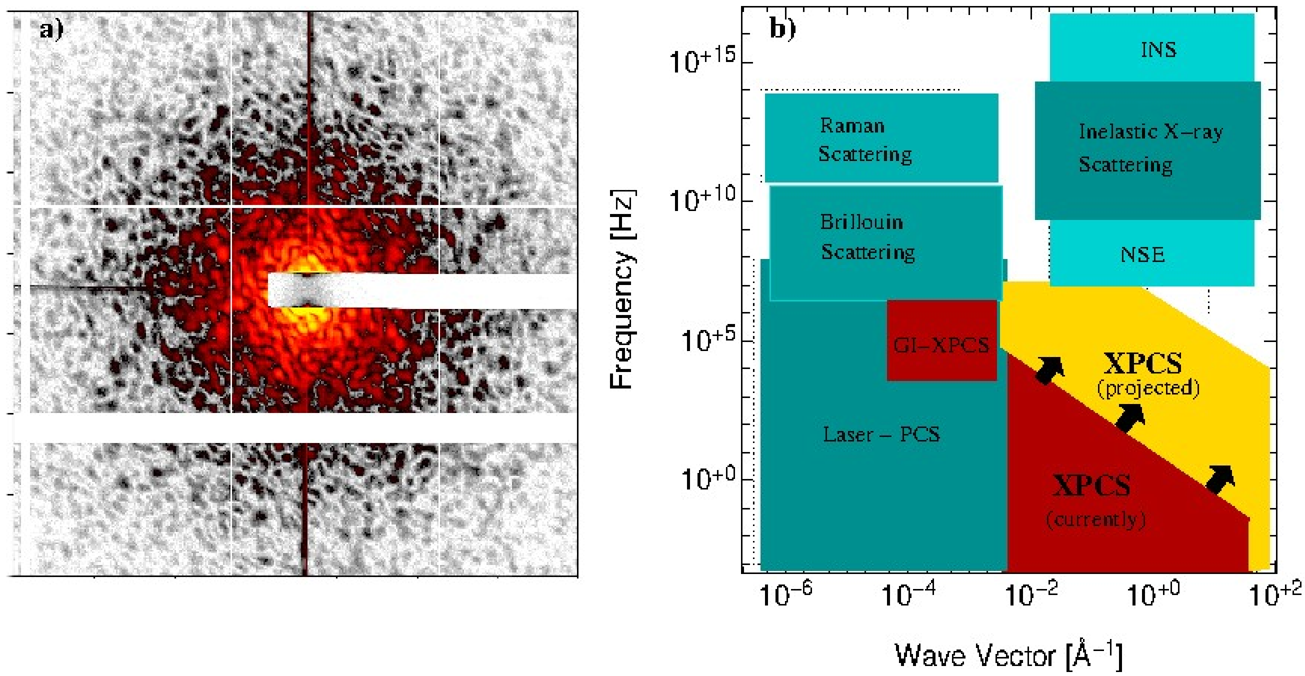

Figure 1 a) Speckle in small-angle X-ray scattering from a nanofabricated sample of Au nanodots on a silicon nitride membrane; b) Wave vector frequency "phase-space" for different experimental techniques measuring the dynamics of materials. With the advent of NSLS-II and other sources such as Petra-III, the capabilities opened by XPCS can now reach faster time scales and shorter length scales than was ever possible before, into a region that is important for understanding the fundamental properties of materials but which is difficult or impossible to explore with other experimental techniques.

The CHX beamline currently uses three Eiger X Pixel Array Detectors (Dectris Inc.). All detectors are equipped with silicon detector chips with 75 x 75 μm^2 pixels. The SAXS detector chamber is equipped with an Eiger 4M detector working up to a speed of 750 Hz with a full dynamic range (12-bit) readout and an Eiger 500K detector capable of continuous operation up to 9 kHz with a 4-bit dynamic range readout. The WAXS detector arm is equipped with an Eiger 1M detector currently operating up to a speed of 3 kHz with a full 12-bit readout dynamic range. Future firmware upgrades from Dectris Inc. are expected to increase the maximum readout frequency of the Eiger 1M detector to 9 kHz and the Eiger 4M to 2.2 kHz with a 4-bit readout. The detectors are permanently installed at the beamline, fully integrated in the control system, and readily available for use.

All the data are collected using *BlueSky* [5] which is Python-based data acquisition software developed by the NSLS-II Data Acquisition and Management (DAMA) group. A majority of the beamline operations conducted by beamline staff and close to 100% of the user operations are based on high level scripts and data acquisition plans developed by DAMA and CHX staff. This results in beamline operations which are efficient even for user groups who possess little or no prior synchrotron experience.

Several analysis methods and algorithms are readily available to the CHX users. Equilibrium dynamics is quantified by intensity autocorrelation functions

$$g^{(2)}(\boldsymbol{q},t)=\frac{\langle I(\boldsymbol{q},t)I(\boldsymbol{q},0)\rangle}{\langle I(\boldsymbol{q},0)\rangle^2}, \tag{1}$$

where I(**q**,t) refers to the initial intensity at the momentum transfer vector ***q***, and time t. The brackets $\langle\ \rangle$ denote ensemble averaging over "initial times" *t=0* and equivalent pixels on the 2D area detector. Out-of-equilibrium and/or non-stationary dynamics XPCS is measured via two-time correlation functions,

$$C(\boldsymbol{q},t_1,t_2)=\frac{\langle I(\boldsymbol{q},t_1)I(\boldsymbol{q},t_2)\rangle}{\langle I(\boldsymbol{q},0)\rangle^2}. \tag{2}$$

Here, the ensemble averages are calculated only over equivalent pixels.

Fourth-order correlation functions $g^{(4)}(\boldsymbol{q},t)$, used to characterize dynamical heterogeneities and capabilities for speckle visibility, are available to access time scales faster than the maximum acquisition rate of the available pixel array detectors when the sample allows it (i.e. if there is sufficient scattering).

The increased flux provides not only access to faster time scales and/or low scattering samples but also creates opportunities for evaluating important effects such as beam damage through a careful analysis of the consistency of measured time scales with different irradiation doses.

All the XPCS analysis methods briefly described above are readily available to the CHX user community through Python-based "analysis pipelines" which are prepared by the beamline staff and available in a public repository [6]. For the time being, the user interface available for XPCS analysis is Jupyter Notebook, an open-source on-line web application supported at NSLS-II by the DAMA group. An example of a Jupyter analysis pipeline can be seen on the CHX GitHub repository site [7].

Considerable efforts have been made to develop complete analysis workflows ("pipelines") for each user experiment, to allow the users to see XPCS analyzed data during the experiment. A "typical" XPCS analysis (e.g. 10^4 frames) may take 10-15 minutes. The analysis is performed on one of the three analysis servers available at the beamline. Typically, two of these servers are reserved for on-line analysis at the beamline (one server for automatic data compression or 'sparsification' and a second one for analysis through the J*upyter* interface). The third server is typically accessed by the entire user community from their home institutions after the beamtime has ended. Once a data analysis is complete, the users can now typically find automatically generated experimental reports attached to the electronic beamline logbook (*Olog*) a mere minutes after data acquisition.

Due to the complexity of the data management and data analysis environment, typically 100% of the users (experts and non-experts alike) require significant support from beamline staff for data analysis while less experienced users also require staff support for XPCS data interpretation. It should also be mentioned that 100% of the data ever recorded at the beamline is analyzed through the mechanism described above. The data is available through the *Databroker* application developed by the DAMA group and linked, through a *MongoDB* database with beamline metadata (beamline state, motor positions, sample information, experimental conditions, state of the storage ring, etc.). Currently, an XPCS experiment uses ~1-2 TB of storage space for the raw data and, depending on the exact analysis being performed, twice this amount for processed data. All the data ever recorded at CHX is currently stored in a ~300 TB Redundant Array of Independent Disks (RAID) storage system installed at the beamline (the amount of disk space is increased periodically). For the longer term, the CHX beamline will require a scalable NSLS-II data storage and data management solution. Various options are being discussed with the DAMA group as part of the future development of the facility.

Beamline Optical Layout

The optical layout of the CHX beamline is schematically shown in Figure 2. X-rays from an in-vacuum undulator with a 20 mm period, IVU20, are conditioned by a horizontal flat mirror and a vertical double-crystal monochromator. (DCM). The cryogenically-cooled DCM is a pseudo channel-cut device designed to maximize the beam stability. The monochromatic x-rays are focused in the vertical direction by a transfocator device using 1-dimensional Be parabolic compound refractive lenses (CRL) (top panel in Figure 2). The horizontal focusing is achieved with a set of parabolic silicon kinoform lenses placed further downstream in the experimental station. With this optical arrangement, a symmetric beam of diameter D ~10 μm is achieved at the sample location. The phase space of the incoming radiation is set to achieve the desired degree of coherence using a set of high-precision monochromatic beam slits placed after the DCM but before the focusing optics. With the SAXS detector placed at a distance of up to 16 m, the angular size of the speckle (λ/D) is sufficient to achieve an oversampling of the speckle if desired. In most cases, the optimal setting for XPCS is a sampling ratio of 1 which maximizes the signal to noise and hence many XPCS SAXS experiments are performed with a detector distance of ~10 m.

The setup for coherent WAXS (Figure 2, lower panel) follows a similar principle. However, to achieve a smaller illumination spot size, both the vertical and the horizontal focusing are done with Si kinoform lenses placed closer to the sample resulting in a higher demagnification ratio. The spot size currently achievable for coherent WAXS is D ~3 μm. With this setup, the WAXS speckle measured with the Eiger 1M detector mounted at 1.5 m are undersampled by a factor of ~0.4, resulting in a corresponding loss of contrast. All Si kinoform optics are developed in house by K. Evans-Lutterodt and tested at CHX as a part of an on-going commissioning program.

In terms of static structures, the available q-range by the SAXS and WAXS instruments covers length scales from 1-10 µm to Ångstroms. For an enhanced longitudinal coherence, the WAXS experiments can benefit from a second set of crystals (Si 220) installed in the DCM. The full c-WAXS capability is still considered as "science commissioning", because important tools such as reciprocal space scanning and mapping and a collision-avoidance protection system are not yet available. However, several general user experiments have already started using the coherent-WAXS capability at the CHX beamline, using angular scans for positioning.

At a storage ring current of 400 mA, the beamline achieves a total coherent flux of about 5×10^{11} photons/s with a 9.6keV partially coherent beam of ~10 µm diameter for coherent SAXS and about 10^{11} photons/s with a 12.8 keV partially coherent beam of ~3 µm diameter for coherent WAXS. The corresponding $g^{(2)}$ contrast factor is ~ 20% for c-SAXS and ~3% for c-WAXS

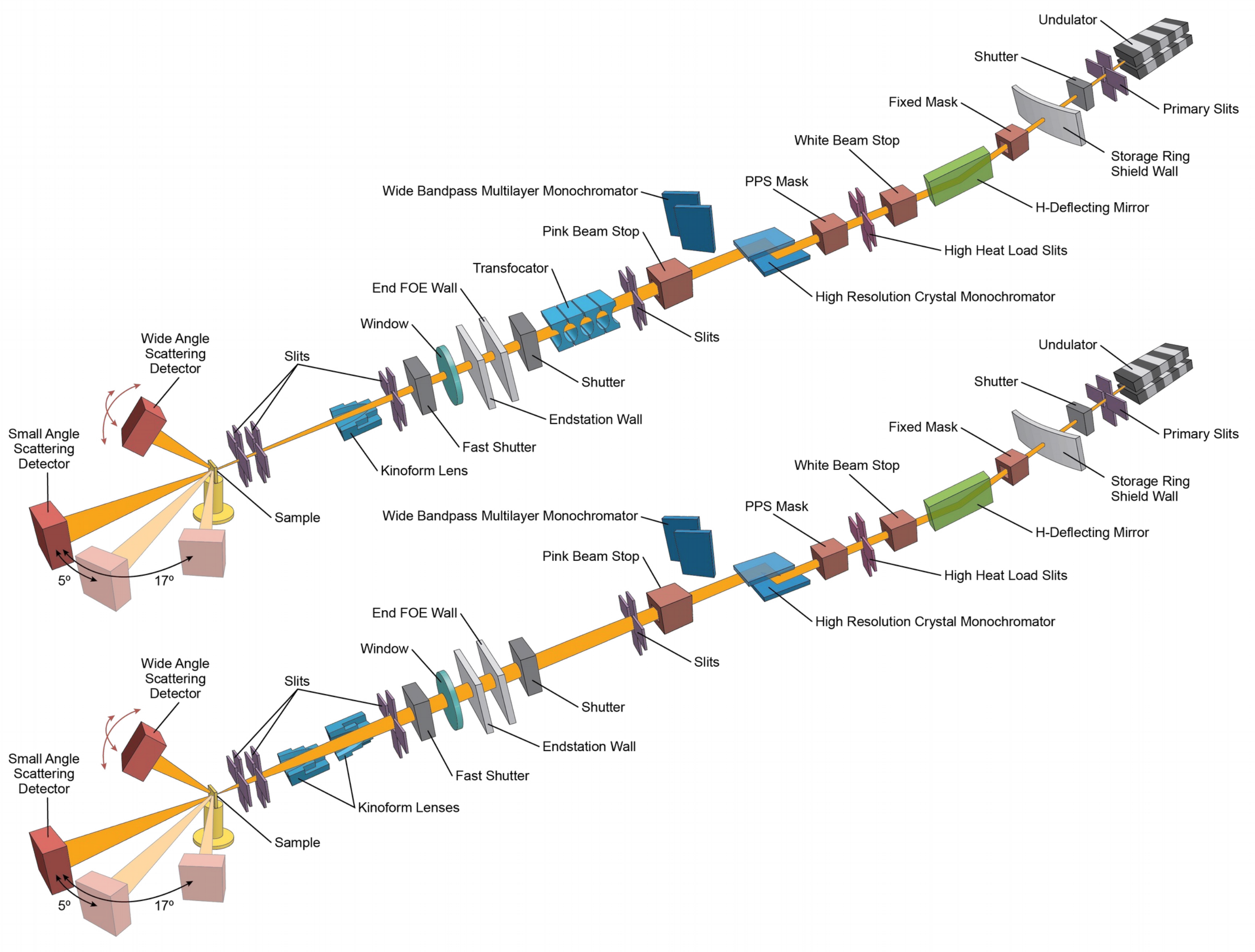

Figure 2 Schematic representation of the CHX optical layout. Top: A focused partially coherent beam of ~10 µm for c-SAXS is obtained with a vertically focused Be CRL transfocator and a set of horizontally focusing Si kinoform lenses. Bottom: a smaller ~3 µm beam for c-WAXS is obtained through a combination of two crossed Si kinoform lenses for vertical and horizontal focusing

Staffing and User Community

Currently, the CHX beamline is supported by the following labor efforts:

- Andrei Fluerasu (1 FTE, lead beamline scientist)
- Lutz Wiegart (1 FTE, beamline scientist)
- Yugang Zhang (~0.75 FTE, scientific associate)

The beamline receives matrixed support through: software engineering (~0.2 FTE), program technician (~0.25 FTE) points of contact for controls and mechanical engineering (~0.2 FTE).

Most of the developments at the beamline are done through strong collaborations with other groups at NSLS-II (Complex Scattering Program, DAMA, Engineering, etc.), BNL (Center for Functional Nanomaterials, Computer Science Initiative) and external collaborators.

A successful user program can flourish only around beamlines where there is a strong in-house research culture and effort. The CHX team is fully dedicated to playing a leading role in the development of XPCS and other related techniques and to establishing an active research program focused on new applications of coherent X-ray scattering.

Fluerasu is collaborating with Prof. Carlos Colosqui from The Mechanical Engineering Department at Stony Brook University on topics related to hydrodynamics, fluid flow at interfaces, imbibition phenomena, electrokinetic flows. Colosqui and Fluerasu have shared a 2016 Joint Photon Sciences Institute Ph.D. research grant and are currently co-supervising the work of a Ph.D. student.

Fluerasu is collaborating with Prof. Matthew Dawber from the Stony Brook Physics Department on applications of XPCS to understanding domain switching dynamics in Ferroelectrics. Dawber and Fluerasu have shared a 2017 BNL-SBU seed grant and are currently co-supervising the graduate work of two Ph.D. students. In 2019, Fluerasu has joined the Physics Department at Stony Brook as an adjunct faculty.

Wiegart is leading a strong NSLS-II effort focused on developing experimental methods and scientific applications of XPCS to direct ink 3D printing. Wiegart is the PI of a 2018 BNL LDRD grant and is currently supervising the post-doctoral work of Dr. Maria Torres-Arango. In his research, Wiegart is collaborating with Profs. T. Koga, D. Gersappe and Y.-C. Chen-Wiegart from the Materials Science Department at Stony Brook University, Dr. Hilmar Koerner from the Air Force Research Laboratory and D. Stanislas Petrash from Henkel Corporation.

Together with Dr. Oleg Chubar from NSLS-II and industrial partners from RadiaSoft Inc. Wiegart is leading an effort focusing on providing advanced simulation tools for the propagation and scattering of partially coherent radiation – i.e. the so-called “virtual CHX beamine”. Wiegart is a co-Insestigator on a 2018 DOE grant led by Chubar.

Zhang is collaborating with the NSLS-II DAMA group and colleagues from APS and, more recently, also ALS on developing a common libraries of advanced tools for XPCS. In his research, Zhang is developing advanced analysis tools such as a multi-tau two-time correlator which has the potential of enabling fast (i.e. real time) analysis for out-of-equilibrium XPCS.

Zhang is collaborating with Prof. Oleg Gang from the CFN and the Mechanical Engineering Department at Columbia University in studying programmed self-assembly in colloidal suspensions and is starting to explore applications of XPCS to the study of the dynamics of self-propelled active colloids. Zhang has accepted general user proposals at the CHX and HXN beamlines

Wiegart, Zhang and Fluerasu are actively collaborating with colleagues from the Complex Scattering Program (M. Fukuto, et al.), the DAMA group and the Computer Science Initiative Department to develop new paradigms for efficient autonomous XPCS data analysis.

Fluerasu, Wiegart and Zhang are active participants and collaborators in a majority of the CHX general user proposals. The CHX beamline users are from diverse scientific disciplines. A majority (57%) of the user experiments are in condensed matter and materials science, about 28% are part of the polymer and soft matter science community and smaller percentages come from life sciences, earth and environmental sciences, instrumentation and optics. Most importantly, we estimate that at least 60% of the CHX users are not coming from the group of "traditional XPCS" users. Indeed, interest in this technique, requiring sophisticated analysis approaches, has extended to an increasingly larger audience.

The ancillary support laboratories were organized in a sub-optimal fashion since the start of operations. CHX shares access to one wet lab and one dry lab with other soft matter and biological beamlines. Both these labs are situated in different LOBs and are consequently only of limited usefulness to beamline staff and users. The situation will greatly improve with the opening of dedicated dry and wet labs in LOB4 in May of this year.

Productivity and Impact

The CHX beamline at NSLS-II provides, together with similar facilities at Petra-III, the highest brightness source (i.e. highest coherent flux) in the 6-16 keV energy range making it thus one of the most attractive instruments worldwide in its category.

The scientific impact of the CHX beamline covers now a broad range of interest from fundamental issues such as the physics of glass transition or dynamics under confinement to industrial applications such as the 3D printing and curing of thermosets. The beamtime usage (beamtime allocated for user experiments) for the CHX beamline has been steadily increasing from ~50% in the 2016-1 cycle to ~80% currently. One of the most important factors that has significantly reduced the beamline productivity during the first two years of operations is a series of facility-wide computer-infrastructure problems which made all data access, data analysis, sometimes even data acquisition all but completely inaccessible during the first half of 2018. While the situation has improved during the last half of 2018, significant managerial decisions and technical developments still need to be made.

A list of notable recent experiments at CHX include:

- Roopali Kukreja *et al*. (U. California Davis), probed dynamics at the crystal-melt interface during the formation of Se nano-crystallites in a binary As-Se glass-forming liquid near its glass transition temperature T g . (submitted 2019)
- Robert Leheny *et al.* (John Hopkins), measured dynamics of nanoparticles under confinement in nanoporous materials (in preparation)
- Y-C Chen-Wiegart *et al.* (Stony Brook U.), mesostructure in 3D printed devices (in preparation)
- H. Koerner *et al.* (Air Force Research Lab.), in-situ monitoring during advanced manufacturing for air force applications, (submitted 2019)
- Tad Koga *et al.* (Stony Brook U.), phase organization (lamellar domains) in block copolymer thin films (main data from CMS, XPCS data from CHX, submitted)
- Randy Headrick, Karl Ludwig *et al.* (U. Vermont, Boston U.), In-situ deposition of thin films, (in review 2019)
- Kevin Yager *et al.* (CFN, BNL), Dynamics of polyelectrolytes under confinement (in review)
- Matt Dawber *et al.* (Stony Brook U.), Dynamics in ferroelectric layered heterostructures (in preparation)
- Bradley Olsen *et al.* (MIT), Dynamics of transient networks of associative polymers

Below we will briefly describe some of the research projects mentioned above, one for each major scattering geometry (SAXS, GI-SAXS, WAXS) and one describing an important in-house effort focusing on developing applications of XPCS to direct ink 3D printing.

Revealing the Dynamics of Polymer Networks by Coherent Small Angle X-ray Scattering

An illustrative example is the data show in Figure 3 recorded from transient networks of associative polymers prepared in the group led by Prof. Bradley Olsen's from MIT. These materials are relevant to artificial skin and self-healing gels and the MIT group is interested in studying their structural dynamics to complement simulation results and visible-light forced-Rayleigh scattering experiments.

Unlike many other similar samples typically studied with XPCS, these gels were not "decorated" with nanoparticles hence the low scattering seen in Figure 3. The Eiger 4M detector was used to record short bursts of 300 frames (=400 ms at 1.33 ms/frame) to mitigate beam damage and the experiments were repeated at fresh locations on the sample to improve statistics.

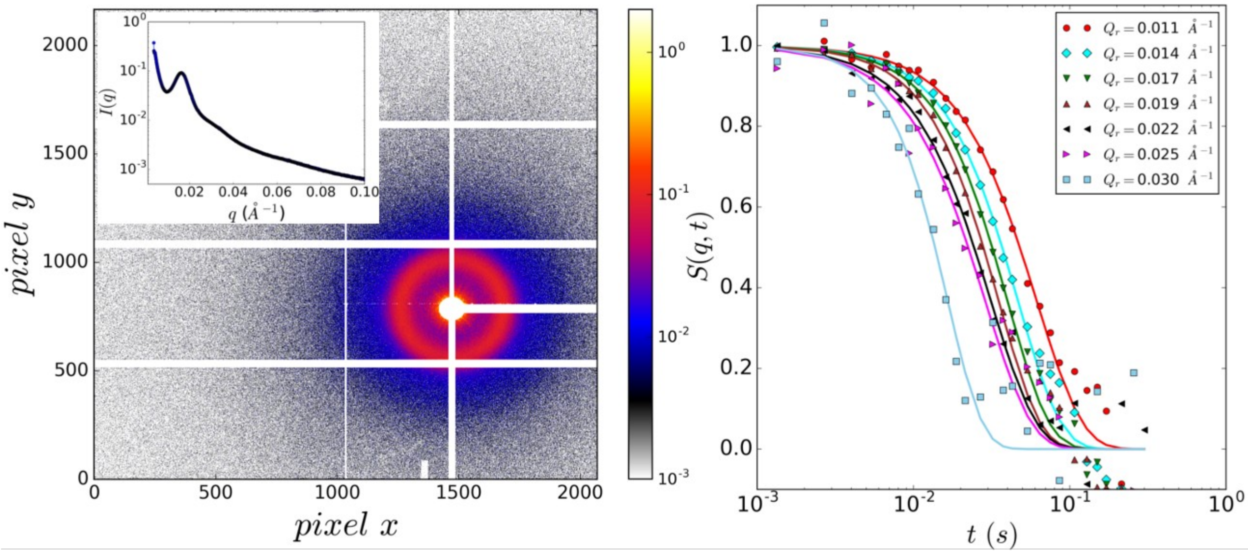

Figure 3 Left: Scattering from transient networks of associative polymerFigure 3 s. The intensity scale on the 2D data recorded with the Eiger 4M detector and radially averaged I(q) shown in the inset is indicated in photons/(frame=1.3 ms) Right: Dynamic structure factor S(q,t) calculated for significantly smaller length scales (q~0.03 Å^{-1}) and shorter time scales (t~10^{-2}s) than previously recorded with similar polymers. B. Olsen et al. (unpublished)

Step-Flow Growth evidenced by coherent GI-SAXS

The CHX beamline is a flexible instrument specifically designed for exceptional stability and versatility. Consequently, the instrument can be adapted to host various in-situ sample handling capabilities such as a rather large custom deposition chamber developed by the group of Prof. R. Headrick from the University of Vermont and his collaborators for studies of thin film growth (Figure 4). In a series of CHX experiments, the team of researchers used XPCS to study vacuum deposition of C_{60} on a graphene-coated surface in surface-sensitive conditions (GI-SAXS). The XPCS data shows evidence of step-flow growth through the observation of the step-edge velocity in the late stages of growth after crystalline mounds have formed.

This research is of a major fundamental and practical importance, providing a better understanding of the properties of artificially grown thin films. Using the CHX capabilities, the team has demonstrated that the use of coherent X-rays provides an approach to understanding aspects such as step flow and other out-of-equilibrium fluctuations which are impossible to access with other techniques.

Unlocking the secrets of domain dynamics in ferroelectric multilayers with coherent WAXS

The research program led by Prof. M. Dawber from Stony Brook University is using coherent-WAXS at CHX to investigate the properties of periodic structures of oxide superlattices where one of the components is a ferroelectric. The chief defining quantity of any ferroelectric material is the existence of a spontaneous polarization below the ferroelectric transition temperature and the formation of spontaneous ferroelectric domains. This research helps advance the understanding of interlayer coupling of domains, and the thermally or applied-field induced motion of those domains, with potentially important implications for the use of these materials in a number of advanced technology and energy applications. In a series of experiments conducted at CHX, various samples containing layers of $PbTiO_3$ (PTO), $SrTiO_3$ (STO) $SrRuO_3$ (SRO) were measured using c-WAXS scattering. Figure 5 shows one of many examples of the thermal and field-induced response/dynamics of such samples.

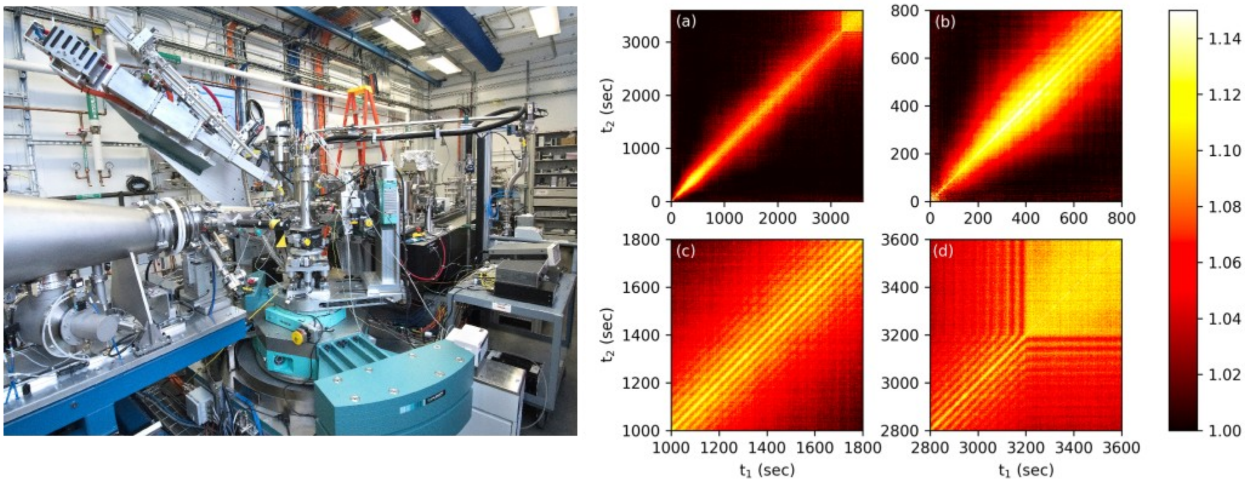

Figure 4 Left: View of the CHX sample area with a custom deposition chamber installed on the horizontal stage of the diffractometer. Right: Sequence of two-time correlation functions showing a complete 1 hour data collection during deposition of C_{60} thin films on graphene substrates at 144°C. The deposition shutter was opened at 40 s. after the start of the scan and closed again at 3200 s. (b) detailed view of the early-time island nucleation and transition to local step-flow growth. (c) close-up during the middle of the scan with stationary dynamics. (d) view of the end of the deposition, showing that the dynamics stop abruptly when the growth shutter is closed. The data was collected at $Q_{\|} = 0.0115$ Å^{-1} and $Q_z = 0.045$ Å^{-1} (see Headrick et al. CHX submitted paper 2)

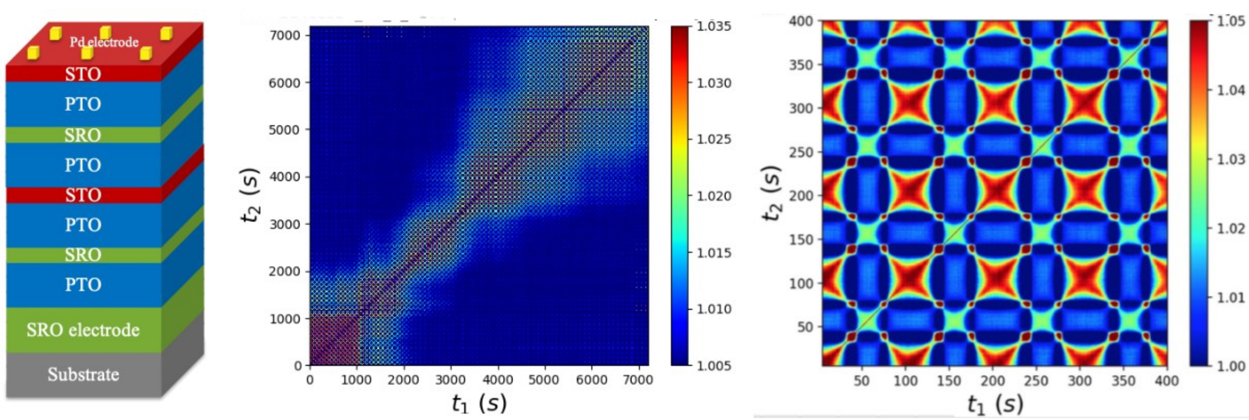

Figure 5: XPCS on layered structures of PTO/STO/SRO hybrid structures (left) held here at 120°C ; The samples are excited by a 5V 10 mHz sine wave for 2hr and the two-time correlation show, in addition to the oscillatory response, a slow, heterogeneous decorrelation by thermal relaxations (middle); A zoom on the two time correlation functions reveal the rich response of the ferroelectric heterostructure to the applied electric field.

In-situ investigations of 3D printing processes

The developments of advanced in-situ sample handling or sample processing capabilities is an important element aiming at enhancing the beamline impact. An illustrative example of a novel sample handling capability is an in-situ platform for direct ink 3D printing developed by a team led by Dr. Lutz Wiegart (CHX). [8] 3D printing is one of the most promising additive manufacturing technologies with potential capabilities ranging from printing automobile, aircraft, and space craft parts to medical (e.g., prosthetics, artificial tissues), optical (e.g., lighting, displays), and microelectronic devices. Given the characteristics of the XPCS technique, the CHX team developed an experimental platform and advanced data handling protocols to study the dynamics of viscoelastic inks during 3D printing processes (see Figure 6). The interplay between nanoscale dynamics and structure is arguably one of the most important factor determining the ultimate macroscopic properties – viscoelasticity, mechanical strength, shape retention, etc. – of the printed material and, from this point of view the CHX beamline can facilitate a fundamental understanding and a better control of such complex manufacturing processes. The CHX 3D printing platform has already successfully been used by general user groups from Stony Brook University, the Air Force Research Lab and industry (Henkel Corporation)

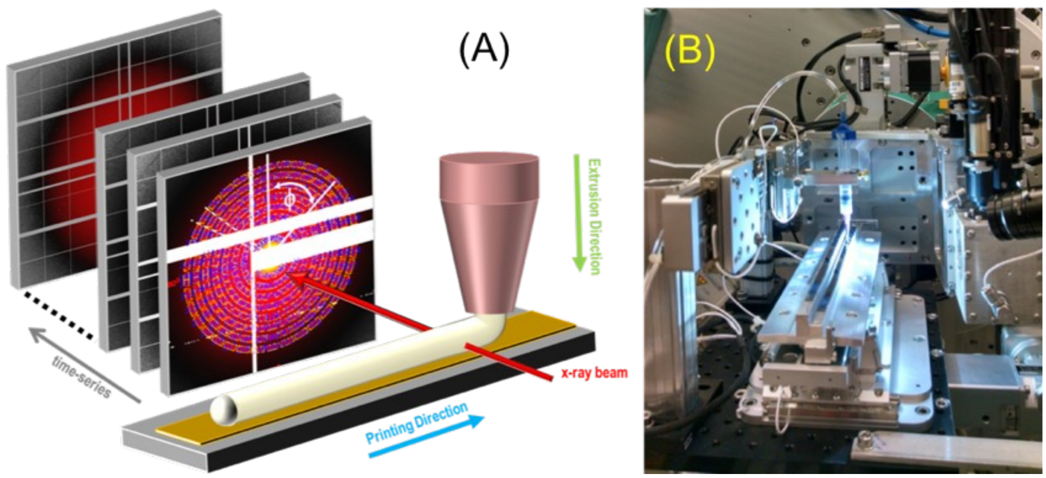

Figure 6: A: Schematic representation of in-situ XPCS experiments during 3D printing processes and B: image of a custom 3D printing platform available to CHX, CMS, SMI users.

Vision for Next Five Years

Over the next period, the CHX beamline will continue to strive for success as a major, world-class instrument for coherent scattering in the medium-hard X-ray regime. Until the startup of new upgraded facilities at ESRF and APS-U, the CHX instrument will continue to provide, alongside Petra-III and Max-IV, the brightest beam in this energy range.

The beamline and NSLS-II staff will continue to make all efforts to improve the data analysis capabilities with the goal of achieving close to real-time data processing and preliminary analysis. The CHX team is currently collaborating with the DAMA group and colleagues from APS and ALS with the goal of establishing a common library for all the major coherent scattering techniques mentioned here – one-time and two-time correlations, 4^{th} order correlations, speckle visibility spectroscopy.

As the amount of collected data increases at a fast rate, it becomes increasingly important to provide capabilities for as much autonomous analysis as possible. To meet this increasing need, the CHX team is collaborating with colleagues from the BNL Computational Science Initiative and other institutions to develop new paradigms for autonomous data analysis based on machine learning.

The beamline staff will continue to develop improved beamline instrumentation. The development of multilayer optics for double multilayer monochromator already existing at the beamline will allow an increase of ~5x in coherent flux for SAXS experiments. The development of better kinoform focusing by e.g. improving the etching processes to increase the working depth of the optics, will continue to remain one of the most important aspects of the commissioning program. The ultimate goal of this effort is to provide diffraction-limited optics for focusing at several energies in the 6-16 keV range (e.g. 6,8,10,12,14,16 keV).

The beamline commissioning/tune-up and the development of new optics will continue to be guided by the rather unique simulation capabilities offered by the so-called "virtual CHX beamline" implemented in the Synchrotron Radiation Software package [9].

Other technical developments which will continue to remain a focus of the CHX beamline staff include, but are not limited to: the implementation of full reciprocal space mapping and reciprocal space scanning capabilities; the development of a robust beamline configuration and collision avoidance management system; the development of various sample handling capabilities such as a high temperature furnace for c-WAXS.

While the development of detectors for XPCS falls outside the scope and technical competence of the CHX staff, we will continue to support efforts such as the development of the VIPIC detector by D.P. Siddons *et al.* It is only through the development of such unique devices that the CHX instrument can achieve its true potential.

The beamline staff will continue to work with vendors such as Dectris Inc. to enable the fastest possible acquisition frame rates on the Eiger detectors and to implement new versions of the Eiger detector firmware.

A successful user program can flourish only around beamlines where there is a strong in-house research culture and effort. The beamline staff will continue to develop existing collaborations with groups from Stony Brook University, the Center for Functional Nanomaterials, The Air Force Research Laboratory and other institutions with the goals of becoming scientific leaders in specific areas of strength:

i. Dynamics and Rheology of Complex Fluids; Providing an improved understanding of the interplay between nanoscale structure and dynamics and macroscopic properties (mechanical, hydrodynamical) in complex materials

ii. XPCS-guided advanced manufacturing (3D printing) and curing technologies

iii. Advanced data analysis tools for coherent X-ray scattering with the goal of approaching real-time autonomous processing through machine learning; Development of advanced new analysis tools (e.g. combined use of XSVS/XPCS to increase the range of accessible timescales, strain mapping, etc)

iv. Domain dynamics in hard materials; Developing tools and methodology for control of nanoscale properties and control of quantum materials

v. Development of advanced simulation tool for coherent wavefront propagation and scattering. Advancing the 'virtual CHX' beamline capability for e.g. efficient feasibility evaluation

The CHX team will continue to work with the entire user community to develop these areas of research while surveying as much of the scientific field as possible for new applications of XPCS and coherent hard X-ray scattering.

It should be pointed out that all these plans may be slowed down to a less than ideal level by insufficient staffing of the NSLS-II beamline(s). To mitigate this situation, the CHX team will continue to seek external funding such as the the JPSI , BNL-SBU seed grant, LDRD and DOE grants awarded to our staff during the last 3 years, and to stay alert for potential new nation-wide research initiatives which may provide opportunities for CHX and the Complex Scattering Program. At the same time, we will continue to advocate for an increased support of this field of research from the NSLS-II and BNL management through concrete measures such as establishing post-doctoral beamline positions and including research in complex soft materials, additive manufacturing, etc. among the laboratory-wide scientific priorities to address the evolving research needs of the nation.

Beamline Self-Assessment

We believe the CHX beamline provides a unique capability to the user community and that the enthusiasm and the efforts of the beamline staff can enable experiments which are currently not possible elsewhere in terms of combination of time- and length- scales and available q-range. The beamline instrumentation is in general reliable with mechanical failures being the exception rather than the rule. In order to become a true world-class facility, the beamline would benefit from more reliable controls and IT infrastructure. By making progress with various automation tasks e.g. through PLC technology, by providing more reliable and more flexible capabilities for user instrument integration in the control system, by fixing important deficiencies in the IT infrastructure (e.g. unreliable file system, non-routable network, lack of basic tools for e.g data transfer, unclear vision about the overall facility data policy) the general user experience could improve significantly, leading to a tremendous increase in the scientific productivity of the facility

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(7) CHX Analysis pipeline https://github.com/NSLS-II-CHX/CHX_Pipelines/blob/master/Beamline_Paper/XPCS_Single_2018_V15_4M_for_uid%3Dc8a1fb1f.ipynb

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Appendix A: CHX Beamline User Statistics

Table A.1 summarizes the CHX Beamline user statistics. The beamtime usage is defined here as the fraction of the beamtime allocated for experiments, which includes general users, science commissioning users, and beamline staff users. Typically, the fraction for beamline staff usage on the level of 10% of the total available experimental time.

The Figures (A1-A3) below show the same beamline statistical data in a graphic format

Table A.1: CHX Beamline user statics by beam cycle.

Beam Cycle	Proposals Submitted	Proposals Allocated	Subscription Rate	Commissioning time (fraction)	Beam Time Usage*	Unique users per CY
2016-1	10	0	N/A	N/A	N/A	
2016-2	25	0	N/A	N/A	N/A	
2016-3	28	6	4.7	0.35	0.65	47
2017-1	29	9	3.2	0.35	0.65	
2017-2	21	12	1.8	0.30	0.70	
2017-3	26	12	2.2	0.28	0.72	34
2018-1	21	16	1.3	0.24	0.76	
2018-2	18	12	1.5	0.22	0.78	
2018-3	24	13	1.8	0.20	0.80	101
2019-1	21	12	1.8	0.20	0.80	
2019-2	22	12	1.8	0.20	0.80	

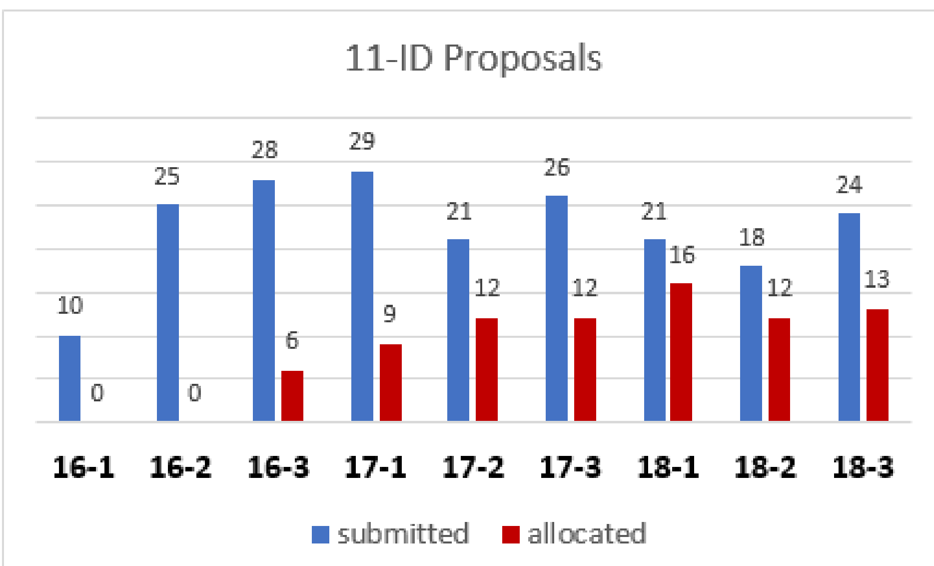
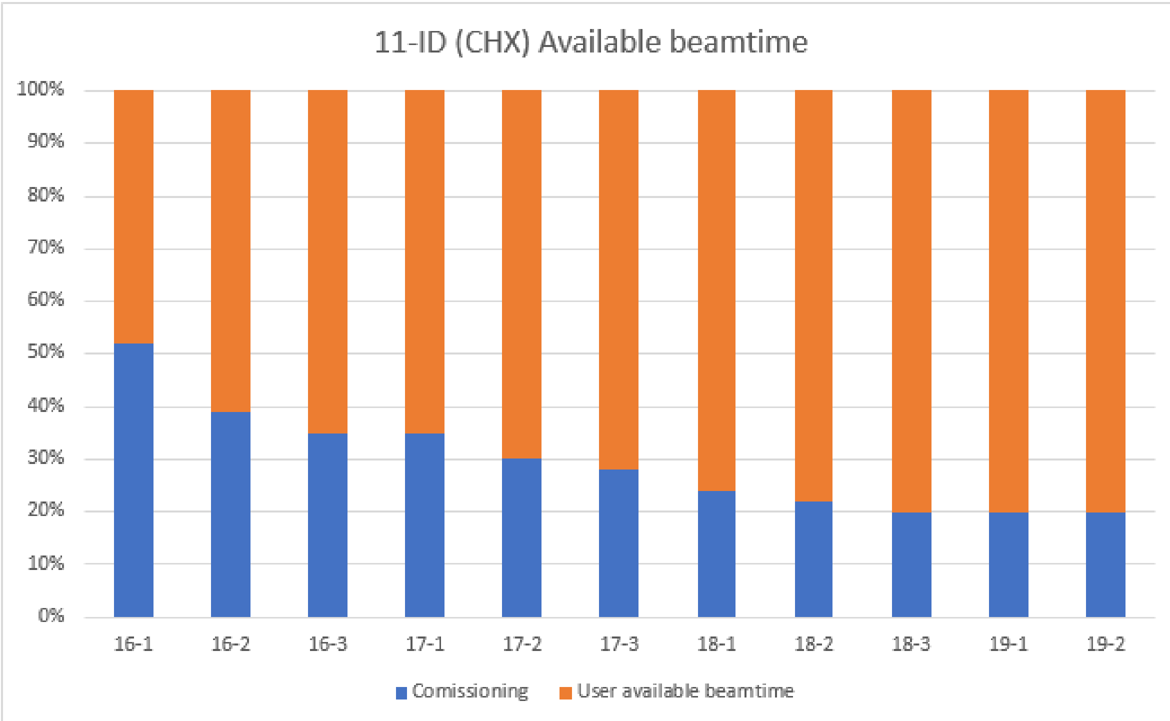


Figure A.1CHX Beamline user statistics;Number of general user proposals and allocated proposals;



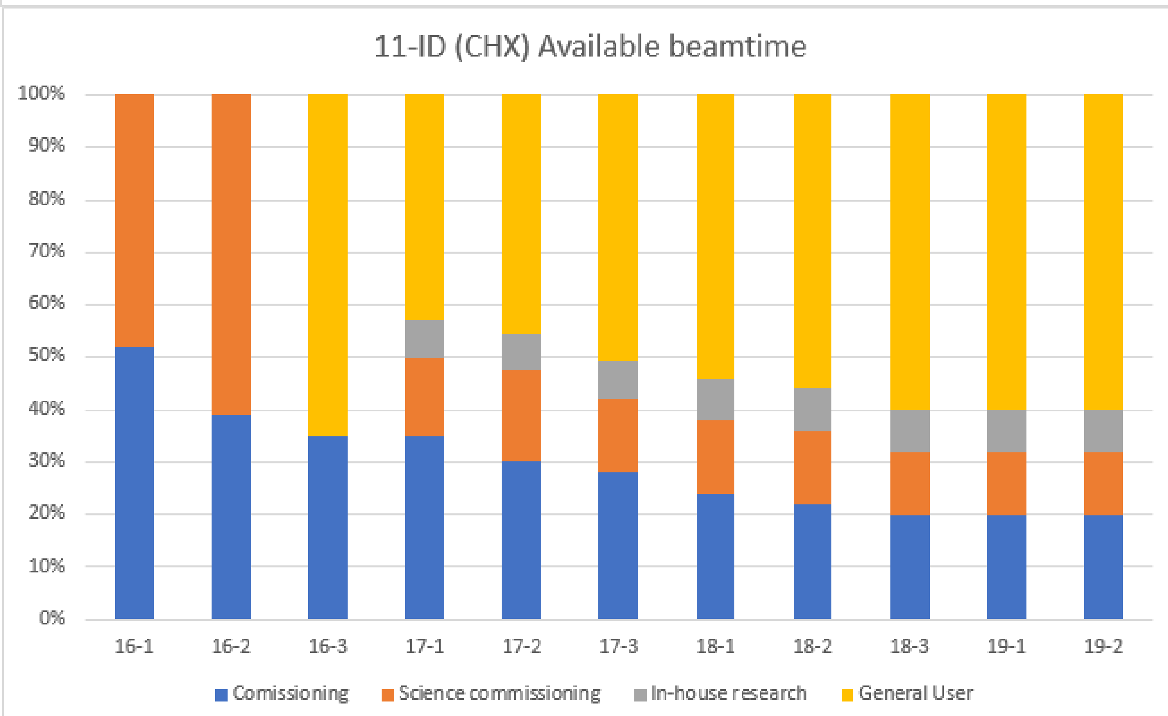


Figure A.2 CHX Beamline user statistics. Top: Percentage of available beamtime usage, defined here as the fraction of the beamtime allocated for experiments, which include general users, science commissioning users, and beamline staff users; Bottom: same as above with detailed representation of general user, science commissioning and beamline discretionary time

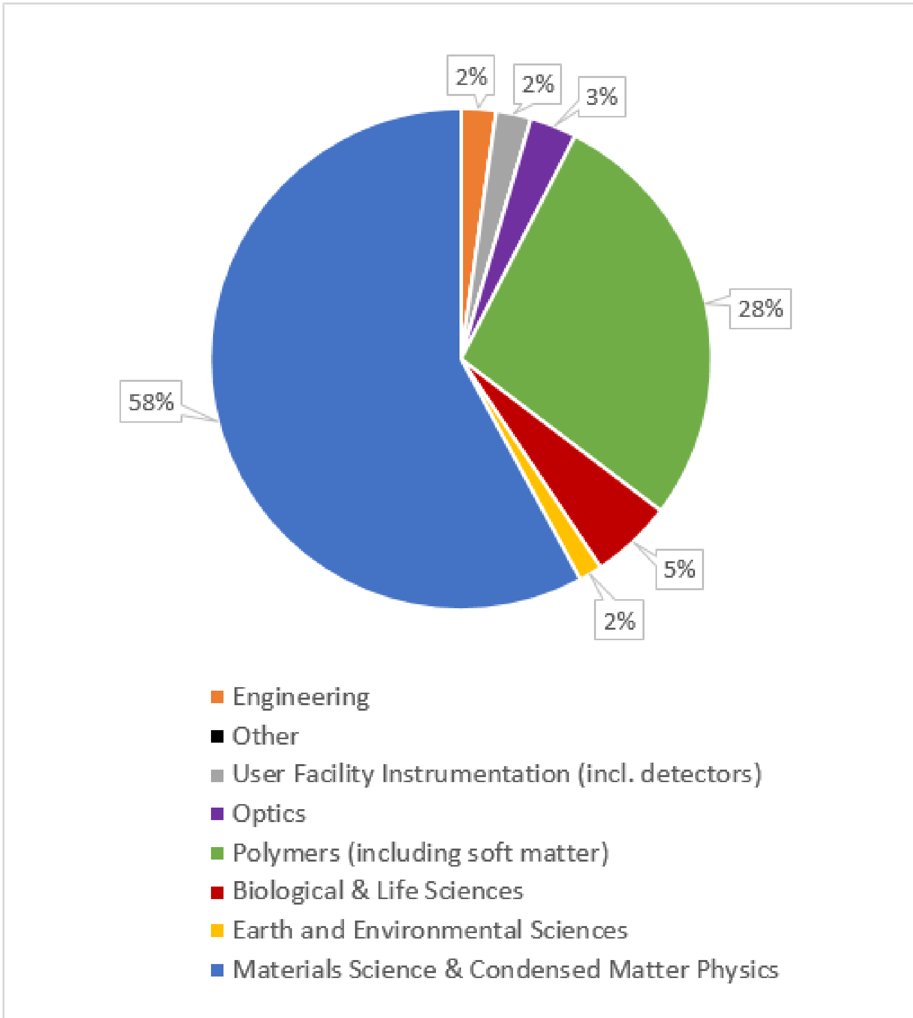


Figure A.3 CHX Beamline user statistics.Distribution of user experiments across various fields

Appendix B: CHX Beamline Publications

Submitted

1. Johnson, Kyle; Wiegart, Lutz; Abbott, Andrew; Johnson, Elias; Baur, Jeffery; Koerner, Hilmar, In operando Monitoring of Dynamic Recovery in 3D Printed Thermoset Nanocomposites by XPCS, Submitted 2019
2. Jianheng Li, Rahul Jangid, Weidi Zhu, Chris Kohne, Andrei Fluerasu, Yugang Zhang, Sabyasachi Sen, Roopali Kukreja, Dynamics at the crystal-melt interface in a supercooled chalcogenide liquid near the glass transition, Submitted 2019
3. Randall L. Headrick, Jeffrey G. Ulbrandt, Peco Myint, Jing Wan, Yang Li, Andrei Fluerasu, Yugang Zhang, Lutz Wiegart, and Karl F. Ludwig, Jr., Coherent X-ray measurement of step-flow propagation during growth on polycrystalline thin film surfaces, in review 2019, https://arxiv.org/abs/1810.11585

<u>Published </u>(papers using CHX X-rays/instrumentation)

1. L. Wiegart, G.S. Doerk, M. Fukuto, S. Lee, R. Li, G. Marom, M. Noack, C. Osuji, M.H. Rafailovich, J. Sethian, Y. Shmueli, M. Torres Arango, K. Toth, K.G. Yager, and R. Pindak, Instrumentation for In situ/Operando X-ray Scattering Studies of Polymer Additive Manufacturing Processes, *Synchrotron Rad. News* **32**, 20 (2019)
2. L Wiegart, M Rakitin, Y Zhang, A Fluerasu, O Chubar, Towards the simulation of partially coherent x-ray scattering experiments, Synchrotron Radiation Instrumentation (SRI 2018), Vol 2054, p. 060079, sponsored by Taiwan Photon Source (2019).
3. Y Choo, P Majewski, M Fukuto, C Osuji, K Yager, Pathway-engineering for highly-aligned block copolymer arrays, Nanoscale, 10, 416-427 (2018).
4. O Ivashkevych, A Munoz, D Poshka, Do you Really Need a Low Current Amplifier to Drive a Low Current Motor?, ICALEPCS 2017, p. 730-733, sponsored by ALBA (2018).
5. O Chubar, C Kitegi, Y Chen-Wiegart, D Hidas, Y Hidaka, T Tanabe, G Williams, J Thieme, T Caswell, et al., Spectrum-Based Alignment of In-Vacuum Undulators in a Low-Emittance Storage Ring, Synch. Rad. News, 31(3), 4-8 (2018).
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8. A Blednykh, B Bacha, G Bassi, W Cheng, O Chubar, A Derbenev, R Lindberg, M Rakitin, V Smalyuk, et al., New aspects of longitudinal instabilities in electron storage rings, Sci. Rep., 8, 11918 (2018).
9. Z Guan, H Qin, K Yager, Y Choo, D Yu, Automatic X-ray Scattering Image Annotation via Double-View Fourier-Bessel Convolutional Networks, British Machine Vision Conference, Vol 2018, p. 1-10, sponsored by British Machine Vision Association (2018).
10. J Lhermitte, A Stein, C Tian, Y Zhang, L Wiegart, A Fluerasu, O Gang, K Yager, Robust X-ray Angular Correlations for the Study of Meso-structures, J. Appl. Cryst., 50, 805-819 (2017).
11. O Chubar, M Rakitin, Y Chen-Wiegart, A Fluerasu, L Wiegart, Simulation of Experiments with Partially Coherent X-rays Using Synchrotron Radiation Workshop, Advances in Computational Methods for X-Ray Optics IV, Vol 10388, sponsored by SPIE (2017).
12. L Wiegart, M Rakitin, A Fluerasu, O Chubar, X-ray Optical Simulations Supporting Advanced Commissioning of the Coherent Hard x-ray Beamline at NSLS-II, SPIE Optical Engineering + Applications, Vol 10388, p. 103880N, sponsored by SPIE (2017).

13. O Chubar, M Rakitin, Y Chen-Wiegart, Y Chu, A Fluerasu, D Hidas, L Wiegart, Main Functions, Recent Updates, and Applications of Synchrotron Radiation Workshop Code , SPIE Optical Engineering + Applications, Vol 10388, p. 1038805, sponsored by SPIE (2017).
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